Int J Thermophys (2010) 31:180–186
DOI 10.1007/s10765-009-0676-1

Determination of Thermal-Diffusivity Dependence on Temperature of Transparent Samples by Thermal Wave Method

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Received: 24 April 2009 / Accepted: 28 October 2009 / Published online: 13 November 2009

Abstract The use of a typical measuring cryostat with a standard temperature controller was proposed for investigation of the temperature dependence of the thermal diffusivity of transparent samples. The basic idea is to use the cryostat heater to control the mean sample temperature and to generate the thermal wave in it, simultaneously. Because of the relatively high thermal inertia of the system, the measurements are carried out at frequencies not exceeding 50 mHz. The periodic temperature disturbance in the sample was detected optically by the use of the mirage effect. The proposed method was used for determination of the thermal diffusivity of yttrium aluminum garnet single crystals in a temperature range from 20 °C to 200 °C.

Keywords Temperature dependence · Thermal diffusivity · Thermal wave measurement · YAG single crystal

1 Introduction

The thermal diffusivity is the main quantity measured in dynamic methods to describe thermal properties of the sample. There are two groups of methods used to determine this parameter: pulse methods and thermal wave methods. The first is based on the generation of temperature gradients by illumination of the sample with a short laser pulse and measurement of the temperature rise versus time on the rear side of the sample. This method is called the flash technique and was introduced by Parker et al. [1] in 1961. It is applied for the characterization of highly conducting materials like metals as well as low conducting polymers and liquids [2–4]. The disadvantage of this method is the necessity of additional sample preparation to obtain discs and, in

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the case of transparent samples, the employment of an additional absorbing layer. The data analysis is based on the determination of the characteristic time in the sample temperature changes.

The other method for determination of the thermal diffusivity is the group of thermal wave methods which are based on the dependence of thermal wave propagation in the sample on its thermal diffusivity. The historically first was the Ångström method developed in 1861 [5]. It was used to characterize the thermal diffusivity of metals by measuring the time delay of the temperature disturbance propagating along a metal rod for two selected points. The idea of Ångström's technique is used in many thermal wave measuring techniques. The periodic temperature disturbance, propagating in the sample as a thermal wave, can be generated in different ways. In photothermal methods the sample is periodically heated by intensity modulated light, which is partially absorbed in the medium. The use of a Peltier module can be also very convenient, especially in investigation of relatively big, transparent samples. In this case, the disturbed temperature field in the sample can be probed by a low power light beam. The probing beam is deflected on a gradient of the refractive index caused by the temperature gradient in the sample (so called mirage effect). This deflection is registered by a position sensor. The mentioned detection method can be very useful for determining thermal properties of either bulk materials or thin films [6,7].

Nowadays electronic and photonic devices often operate at elevated temperature. Thus, the design of these devices requires a knowledge of material properties as functions of the temperature. The dependence of the thermal diffusivity on the temperature can be determined by the flash technique over a wide temperature range. However, in the case of transparent samples the necessity of application of additional opaque films deposited on the sample surfaces may lead to systematic errors during the experiment. The results of our earlier investigations seem to confirm this conclusion [8,9]. In this work, we present a variant of the measuring technique used for investigation of transparent single crystals. It allowed determination of the thermal diffusivity of transparent bulk materials as a function of temperature. In the next section the experimental setup is described. Then results of measurements carried out for a YAG single crystal are presented.

2 Experimental Technique

As was mentioned in the introduction, the measuring method described in this paper is the variant of a method used for determination of the thermal diffusivity of transparent samples. In the previous case, the thermal wave propagating in the sample was generated by a Peltier element [10]. The modification consisted in the placement of the investigated sample in the cryostat (VPF-700 from JANIS). One surface of the sample was in good thermal contact with a copper plate called a "cold finger," as a result of the use of silicon paste (Fig. 1). The cryostat chamber was evacuated to a pressure of a few Pa. It assured good thermal insulation of all sample walls except for that contacting the cold finger. The cold finger temperature was controlled by the temperature controller (Lake Shore Controller 331). At the beginning of a measurement, the sample temperature was stabilized at a set temperature. Then the periodic temperature

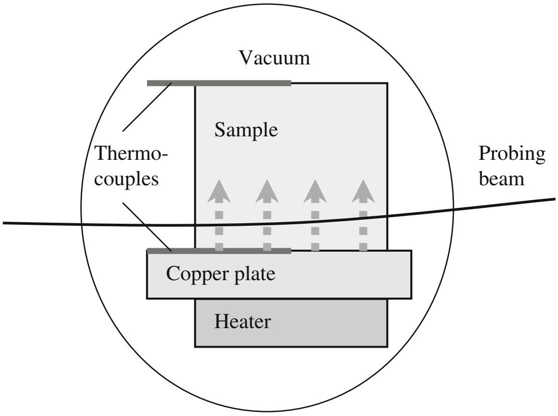

Fig. 1 Geometry of the experiment

modulation around the set point was applied. As an effect, the cold finger periodically heated and cooled the one sample surface with a constant frequency. Because of the thermal inertia of the system, the modulation frequency did not exceed 50 mHz. The temperature in the system was measured by two thermocouples, one placed on the upper side of the sample, and the other placed on the cold finger. It allowed control of the actual temperature of the sample.

The temperature disturbance propagated in the sample was probed by a light beam from a He–Ne laser (Lasos 7672). The deflection of this beam was detected by a position sensing diode (DL400-7PCBA) and then, its amplitude and phase were measured by a DSP lock-in amplifier (EG&G 7265) referred to the modulation frequency provided by the temperature controller. The amplitude and the phase were determined as functions of the distance of the probing beam from the heated sample surface. The experimental results were collected on a PC via a GPIB interface.

In the experiment, the thermal diffusivity was determined on the basis of a one-dimensional (1D) model of the thermal wave propagation in the sample [11]. The density of the heat flux introduced to the sample is described by the formula,

$$j(x = 0) = A\cos(2\pi ft) \tag{1}$$

where A is its amplitude and f is the modulation frequency. The measured signal is proportional to the temperature gradient existing in the sample and can be written as

$$S(x,t) \sim \frac{\partial T(x,t)}{\partial x}. \tag{2}$$

Solving the Fourier–Kirchhoff equation,

$$\frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial x^2} \tag{3}$$

with the boundary conditions,

$$-\alpha C\frac{\partial T}{\partial x}\bigg|_{x=0} = A\exp(\mathrm{i}2\pi f t)\,, \quad -\alpha C\,\frac{\partial T}{\partial x}\bigg|_{x=d} = 0 \tag{4}$$

where α is the thermal diffusivity of the sample, C is the heat capacity, and d is the sample thickness, one can get the formula describing the temperature field in the sample:

$$T(x,t) = \frac{A}{\kappa}\sqrt{\frac{\alpha}{\mathrm{i}2\pi f}}\,\frac{\sinh\left[\sqrt{\frac{\mathrm{i}2\pi f}{\alpha}}\,(d-x)\right]}{\sinh\left(\sqrt{\frac{\mathrm{i}2\pi f}{\alpha}}d\right)}\exp(\mathrm{i}2\pi f t) \tag{5}$$

In the case of thermally thick samples, the formula for a thermal wave propagating in the sample can be simplified and written as

$$T(x,t) = T_0\exp\left(-\sqrt{\frac{\pi f}{\alpha}}x\right)\cos\left(2\pi f t - \sqrt{\frac{\pi f}{\alpha}}x + \varphi_0\right) \tag{6}$$

where T_0 is the amplitude of the thermal wave and φ_0 is a constant. The phase delay of the deflected signal is described by the expression

$$\Delta\varphi = -\sqrt{\frac{\pi f}{\alpha}}\Delta x + \varphi_0 \tag{7}$$

According to Eq. 7, the thermal diffusivity was evaluated from fitting the experimental data with a straight line.

3 Results and Discussion

The measurements were provided for a few YAG ($Y_3Al_5O_{12}$) single crystals grown by the Czochralski technique. The samples were oriented along the main crystallographic directions $[1\bar{1}0]$, [111], and [224]. The thermal diffusivity was determined over a temperature range from 20 °C to 200 °C. As was mentioned in Sect. 2, the cold finger periodically heated and cooled the sample surface. The temperatures of the cold finger and this upper surface were measured by thermocouples. The signals from the bottom and upper thermocouples are shown in Fig. 2. There is a clear shift between these two signals connected with propagation of the thermal wave in the sample. The experiment started after achieving appropriate modulation of the temperature.

For each temperature the phase delay and the amplitude of the deflected signal were measured. Because of instability of the signal amplitude during the experiment corresponding to unstable work of the heating element, the analysis of experimental data concerned only the phase delay of the signal. Referring to Eq. 7, the phase delay of the signal is a linear function of the position of the probing beam in the sample. Thus,

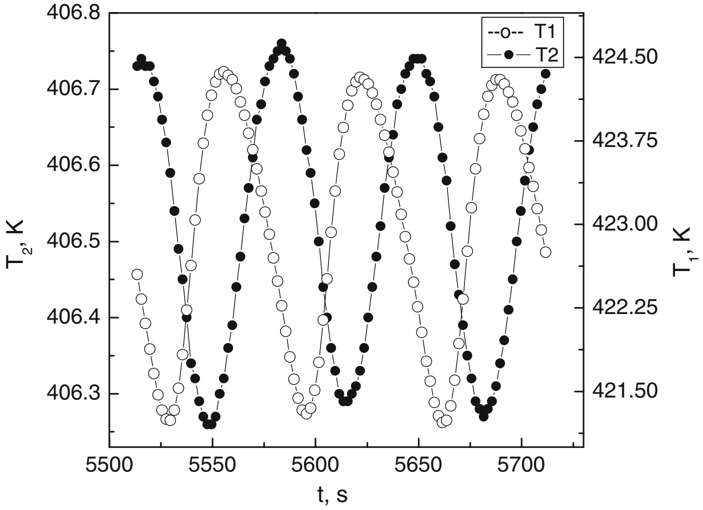

Fig. 2 Signals from thermocouples

Table 1 Thermal diffusivity results

Sample	Thermal diffusivity ($10^{-2}\text{cm}^2 \cdot \text{s}^{-1}$)			
	22 °C	132 °C	150 °C	182 °C
YAG	4.46(52)	3.88(45)	2.45(57)	1.29(74)

the experimental data were fitted with a straight line (Fig. 3a). The thermal diffusivity was determined from the slope coefficient a according to the equation,

$$\alpha = \frac{\pi f}{a^2} \tag{8}$$

The modulation frequency was equal to $f = 20\,\text{mHz}$. The thermal-diffusivity results are reported in Table 1 and shown graphically in Fig. 3b. The thermal diffusivity decreases with an increase in the temperature. The value determined at 182 °C is almost 30 % smaller than that determined at room temperature. The results for a typical YAG laser crystal correspond to literature values [12].

4 Conclusion

A variant of the thermal wave method was used to provide measurements of the thermal diffusivity of transparent samples as a function of temperature. The generating beam typically used in the photothermal experiment was replaced by an electrical heater controlled by a temperature controller, and the sample was placed in the cryostat chamber for better thermal insulation. The results showed that this is a very convenient technique allowing determination of the temperature dependent thermal diffusivity in a

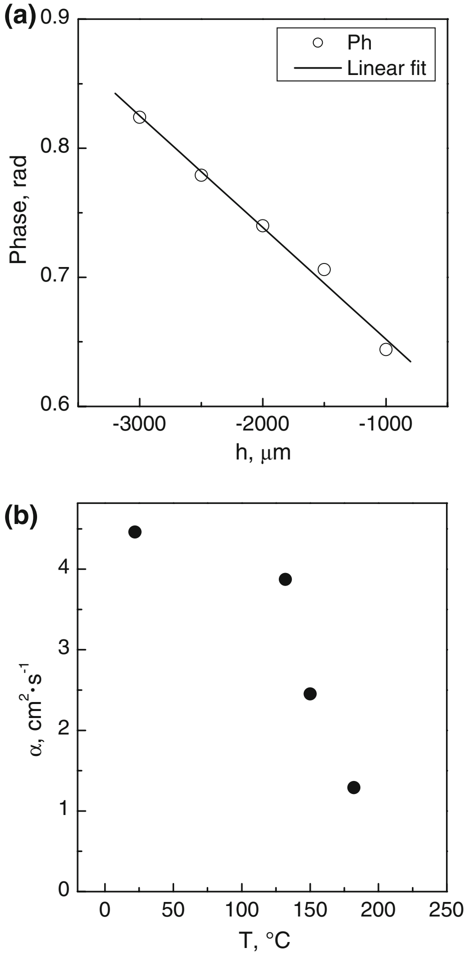

Fig. 3 (**a**) PD signal measured as a function of the distance between the probing beam and a heated sample surface for a temperature of 132 °C and (**b**) thermal-diffusivity results for pure YAG crystal

non-contact and non-destructive way. The proposed setup configuration is dedicated to characterization of thermal properties in a wide range of temperature from room temperature to a few hundred kelvin without the necessity of special sample preparation. According to the optical signal detection, the sample should be of good optical quality and without macroscopic defects of its structure. The signal is measured in the sample and carries direct information about the thermal properties. Experimental results are in good agreement with literature values.

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